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(54) Title: INTEGRATED PROCESS FOR THE PRODUCTION OF OLEFIN OXIDES

(57) Abstract: Integrated process for the production of olefin oxides in which a dehydrogenation unit, a hydrogen peroxide synthesis unit and an epoxidation unit of C₂-C₅ olefins are integrated with each other and wherein the hydrogen coming from the dehydrogenation forms a raw material for the preparation of hydrogen peroxide which is fed to the epoxidation unit together with the olefin produced.

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INTEGRATED PROCESS FOR THE PRODUCTION OF OLEFIN OXIDES

The present invention relates to an integrated process for the production of olefin oxides.

10 More specifically, the present invention relates to an integrated process for the production of olefin oxides starting from a C₂-C₅ alkyl stream.

Even more specifically, the present invention relates to an integrated process for the production of propylene
15 oxide from propane, by the direct oxidation of propylene with hydrogen peroxide.

Olefin oxides, or epoxides, are useful intermediates for the preparation of a wide variety of compounds. For example, olefin oxides can be used for the production of glycols, condensation polymers such as polyol polyethers or
20 polyesters which can be used as such or as useful intermediates in the synthesis of polyurethane resins, elastomers, sealants, etc.

The preparation of epoxides by the direction oxidation
25 of the corresponding olefins with hydrogen peroxide in the

presence of catalysts such as titanium-silicalite treated with neutralizing substances to neutralize the acid groups present on the surface, is known in literature. Catalysts of this type are described, for example, in European patents EP 230,949, EP 712,852 and EP 940,393 or in published international patent application WO 00/17,178.

In literature, the synthesis of epoxides is therefore described starting from the assumption that the reagents, essentially olefin and hydrogen peroxide, are available as already preformed streams. On an industrial scale, however, the availability of raw materials for the preparation of a product such as an olefin oxide is not always immediate or is not always economically advantageous for the market value of the epoxide.

To solve the problem of the availability of raw materials, the Applicant has now found a process for the production of olefin oxides in which the preparation of the desired epoxide is integrated with that of the olefin and hydrogen peroxide so that the final synthesis depends exclusively on easily available raw materials such as oxygen and the corresponding alkyl hydrocarbon.

The object of the present invention therefore relates to an integrated process for the production of olefin oxides which comprises:

a) feeding in continuous to a dehydrogenation unit, a par-

affinic stream essentially consisting of at least one C₂-C₅ alkyl hydrocarbon;

- b) sending the stream leaving the dehydrogenation unit to a separation unit to produce a stream consisting of hydrogen, a stream essentially consisting of the non-reacted alkyl hydrocarbon, which is recycled to the dehydrogenation, and a stream essentially consisting of the dehydrogenated alkyl hydrocarbon (olefin stream);
- c) feeding the stream of hydrogen together with a stream of oxygen, to a hydrogen peroxide synthesis unit;
- d) feeding a part of the olefin stream, together with the hydrogen peroxide produced in step (c), to an oxidation unit, the rest of the olefin stream being sent to a storage tank;
- e) recovering the olefin oxide produced and sending it for storage.

According to the present invention, the paraffinic stream fed to the dehydrogenation can be selected from one or more of the C₂-C₅ alkyl hydrocarbons even if it is preferable to operate with a stream of ethane and/or propane or, even more preferably, with a stream of propane alone.

The paraffinic stream, coming from a refinery with a purity degree higher than or equal to 95%, is fed to the dehydrogenation unit which effects a catalytic dehydrogenation at a temperature ranging from 450 to 800°C, preferably

from 550 to 650°C, and at an absolute pressure ranging from 10 to 300 kPa. Any dehydrogenation catalyst for light paraffins can be used in the process, object of the present invention, even if catalysts based on platinum, gallium, chromium, vanadium supported on silica and/or alumina are preferred. Details on the dehydrogenation of paraffins and on the catalysts used are available on SRI International "Alkane Dehydrogenation and Aromatization", Report 203, September 1992.

At the end of the dehydrogenation, the stream produced is sent to a two-step separation unit. In the first step, the hydrogen is recovered whereas in the second step the non-reacted paraffin is separated from the olefin.

The separation of the hydrogen also generally takes place in two steps. In the first step, carried out with a distillation system of the cold box type, a stream of hydrogen is recovered, still containing significant quantities of impurities, whereas in the second step a purification is effected, for example by means of membrane separation or PSA (Pressure Swing Adsorption), which allows a stream of hydrogen to be obtained with a purity suitable for the subsequent hydrogen peroxide synthesis reaction.

The hydrogen, together with a stream of oxygen, is fed to a hydrogen peroxide synthesis unit which operates with well-defined operating conditions. In particular, the hy-

hydrogen peroxide can be prepared by feeding in continuous to a stirred reactor containing, in dispersion, a heterogeneous catalyst based on platinum and palladium, a stream consisting of:

- 5 - a liquid stream containing an alcohol or an alcohol-water mixture with an alcohol content higher than 50% by weight, and an acid or halogenated promoter; and
- a gaseous stream containing hydrogen, oxygen and an inert gas, wherein the concentration of hydrogen is lower than 4.5% by volume and the concentration of oxygen is 10 lower than 21% by volume, the complement to 100 being the inert gas, for example nitrogen or a noble gas such as helium or argon.

A liquid stream is extracted in continuous from the 15 synthesis reactor, containing the hydrogen peroxide produced in a concentration ranging from 2 to 10% by weight and a gaseous stream essentially consisting of non-reacted hydrogen and oxygen and the inert gas, which is recycled.

The synthesis reaction is carried out at a temperature 20 ranging from -10 to 60°C, preferably from 0 to 40°C, at a pressure ranging from 1 to 300 bars, preferably from 40 to 150 bars, and with residence times of the liquid medium in the reactor ranging from 0.05 to 5 hours, preferably from 0.1 to 2 hours.

25 The hydrogen peroxide synthesis catalyst contains pal-

ladium in a quantity ranging from 0.1 to 3% by weight and platinum in a quantity ranging from 0.01 to 1% by weight, with an atomic ratio between platinum and palladium ranging from 1/500 to 100/100. The palladium is preferably present in a quantity ranging from 0.4 to 2% by weight and the platinum in a quantity ranging from 0.02 to 0.5% by weight, with an atomic ratio between platinum and palladium ranging from 1/200 to 20/100. In addition to the two metals mentioned above, the catalytic system can comprise one or more promoters selected from metals of groups VIII or IB, such as ruthenium, rhodium, iridium and gold, in a concentration generally not higher than that of the palladium.

The catalyst can be prepared by dispersing the active components on an inert carrier by means of precipitation and/or impregnation starting from precursors, for example salts or soluble complexes. The inert carrier can be selected from silica, alumina, silica-alumina, zeolites, activated carbon. Activated carbon with a surface area ranging from 300 to 1400 m²/g is preferred.

The catalyst is normally dispersed in the reaction medium at a concentration ranging from 0.1 to 10% by weight, preferably from 0.3 to 3% by weight.

The liquid stream consists of one or more C₁-C₄ alcohols or a mixture of these alcohols with water. Among the alcohols, methanol is preferred.

The acid promoter can be any substance capable of generating hydrogen ions in the liquid reaction medium and is generally selected from inorganic acids such as sulfuric, phosphoric, nitric acids or from organic acids such as sulfonic acids. Sulfuric acid and phosphoric acid are preferred.

The concentration of the acid generally ranges from 20 to 1000 mg per kg of liquid medium and preferably from 50 to 500 mg per kg of liquid medium.

10 The halogenated promoter can consist of any substance capable of generating halogen ions in the liquid reaction medium. These substances are generally capable of generating bromide ions such as hydrobromic acid and its salts soluble in the reaction medium, for example alkaline bromides. The concentration of halogenated promoter generally ranges from 0.1 to 50 mg per kg of liquid medium and preferably from 1 to 10 mg per kg of liquid medium.

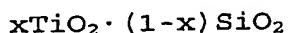
20 The reaction product containing hydrogen peroxide, after filtration for recovering the catalyst dispersed therein, is fed directly to the epoxidation unit of the olefin together with the latter. In order to keep the material balance under conditions of self-sufficiency, the fraction of olefin capable of being oxidized by the hydrogen peroxide produced with the hydrogen coming from the dehydrogenation unit, is fed to the epoxidation unit. The re-

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maining olefin is sent for storage.

The reagent streams are fed to the epoxidation reactor under such conditions as to have a molar ratio olefin/H₂O₂ inside the reaction container, ranging from 10/1 to 1/10, preferably from 6/1 to 1/1. The epoxidation reaction is carried out under stirring, in the presence of the same solvent used in the hydrogen peroxide synthesis, at a pH ranging from 5.5 to 8 and in the presence of a catalyst used in a quantity ranging from 1 to 15% by weight with respect to the reaction mixture, preferably from 4 to 10%.

Any epoxidation catalyst can be used in the process, object of the present invention even if titanium-silicalite having the general formula:



is preferred, wherein x represents a number ranging from 0.0001 to 0.04, preferably from 0.01 to 0.025. This catalyst is described in the patents U.S. 4,410,501, 4,824,976, 4,666,692, 4,656,016, 4,859,785, 4,937,216.

The epoxidation reaction of the olefin is carried out at a temperature ranging from 20 to 150°C, preferably from 40 to 100°C, even more preferably from 55 to 90°C and a such a pressure as to keep the olefin dissolved in liquid phase at the reaction temperature.

The stream leaving the epoxidation reactor is sent to a purification section from which the olefin oxide produced

is recovered and sent for storage, together with the solvent which is recycled both to the hydrogen peroxide synthesis reactor and to the oxidation reactor of the olefin.

In the process, object of the present invention, the most critical section is the separation section of the olefin from the corresponding alkyl hydrocarbon as the products to be separated, for example ethane/ethylene or propane/propylene have very similar boiling points and also because the separation may require the use of a cryogenic unit whose running involves problems of a technological/economic nature. To reduce the incidence of this problem, it is possible, according to the present invention, to use the epoxidation unit as an additional partial separation section of the hydrocarbon mixture leaving the dehydrogenation unit.

A further object of the present invention therefore relates to an integrated process for the production of olefin oxides which comprises:

- a) feeding in continuous to a dehydrogenation unit, a paraffinic stream essentially consisting of at least one C_2 - C_5 alkyl hydrocarbon;
- b) sending the stream leaving the dehydrogenation unit to a first separation unit to produce a stream consisting of hydrogen and a mixed stream essentially consisting of the non-reacted alkyl hydrocarbon and the dehydrogenated

alkyl hydrocarbon (olefin);

c) feeding the stream of hydrogen together with a stream of oxygen, to a hydrogen peroxide synthesis unit;

d) feeding a part of the hydrocarbon stream coming from the first separation section, together with the hydrogen peroxide produced in step (c), to an oxidation unit;

e) feeding the stream leaving the epoxidation unit to a second separation unit to recover a stream containing the olefin oxide produced and a stream essentially consisting of the non-dehydrogenated alkyl hydrocarbon which is recycled to the dehydrogenation;

f) feeding the remaining part of the hydrocarbon stream coming from the first separation section to a third separation section to recover a paraffinic stream, recycled to the dehydrogenation, and an olefin stream sent for storage.

Also in the additional process, object of the present invention, in order to keep the material balance under conditions of self-sufficiency, a fraction of mixed hydrocarbon stream which contains a quantity of olefin capable of being oxidized by the hydrogen peroxide produced with the hydrogen coming from the dehydrogenation unit, is fed to the epoxidation unit.

The process, object of the present invention, can be better described by referring to the drawings of the en-

closed figures which represent an illustrative but non-limiting embodiments, wherein:

Figure 1 represents a block scheme of the present process wherein a fraction of the olefin produced after the dehydrogenation phase is fed to the epoxidation unit;
5 and

Figure 2 represents a block scheme of the present process wherein a fraction of the mixed stream still containing both the olefin and non-dehydrogenated corresponding paraffin is fed to the epoxidation unit.
10

With reference to the figures, D represents the dehydrogenation unit, H represents the hydrogen peroxide synthesis unit, E represents the epoxidation unit, S1-S3 are three separation units, P represents a purification unit of the hydrogen stream, R is a recovery unit of the solvent
15 whereas PR and PO represent the storage tanks of the olefin and epoxide respectively.

The integrated process for the production of olefin oxides, object of the present invention is evident from the enclosed figures and following description. In particular,
20 a paraffinic stream coming from a refinery, for example propane with a purity of over 98%, is fed by means of line (1) to the dehydrogenation unit D. The outgoing stream is subjected to a first separation, for example in a distillation system of the cold box type S1, to recover the hydro-
25

gen (2) which, after purification in P (membrane separation or PSA), is fed, by means of line (3), to the hydrogen peroxide synthesis unit H together with the stream of oxygen and inert gas (9).

5 The stream (5) leaving S1 goes to the separation section S2, to recover the non-dehydrogenated paraffin (6), which is recycled to the unit D, and the olefin (8) which, according to the scheme of figure 1, is partly stored in PR and is partly sent, by means of line (7), to the oxidation
10 unit E. The light products or impurities (methane, ethane, ethylene, heavy products), which are discharged from sections S1, S2 and P are sent to the torch by means of line (4).

 The stream (14) containing hydrogen peroxide in solution is fed to the oxidation unit E, together, as already
15 mentioned, with the olefin (7). The solvent is recovered in the unit E from the stream (11), which is separated in R and recycled both to the hydrogen peroxide synthesis, by means of (12), and to the oxidation unit, by means of (13),
20 as diluent of the hydrogen peroxide solution (10) leaving the synthesis. The olefin oxide produced (16) goes to the separation/purification section S3 from which the non-reacted olefin (17) is recovered, which is recycled to E, and the epoxide (18) stored in PO.

25 The excess water formed during the cycle is separated

from the solvent in R, treated with means not illustrated in the figures, and discharged to the sewage by means of line (15).

In the variations of Figure 2, the hydrocarbon stream (7) is fed to the oxidation unit. In particular, the stream (5) leaving the separation unit S1 is partially fed to the oxidation unit E, by means of (7), and partially to the separation section S. The reaction product (16) leaving unit E is sent to the separation/purification section S3 from which, in addition to the epoxide, the stream (17) containing the non-dehydrogenation paraffin and the non-reacted olefin in E, is recovered. This stream is recycled to the dehydrogenation unit D.

An illustrative but non-limiting example is provided for a better understanding of the present invention and for its embodiment.

EXAMPLE

The procedure is adopted according to the scheme of Figure 1. The dehydrogenation step is carried out using a fixed bed reactor and a chromium catalyst on alumina. The dehydrogenation temperature is 650°C, the pressure 70 kPa.

The hydrogen peroxide synthesis is effected in a solvent (methanol) in which a catalyst consisting of palladium and platinum on coal, is dispersed at a concentration of 1% by weight. 200 mg of H_2SO_4 per kg of liquid medium and 6

mg of HBr per kg of liquid medium, are used as reaction promoters. The reaction temperature is 25°C, the pressure is 10 MPa. A hydrogen peroxide solution at 7% is obtained, which, after dilution to 3.5%, is fed to the epoxidation unit.

The latter is effected using titanium silicalite dispersed in a concentration of 6% by weight in the aqueous reaction medium kept at a pH of 6.5 by the addition of NH_4OH . The reaction temperature is 50°C, the pressure 1.3 MPa.

The material balances are indicated in the following table.

Component	Stream											
	1	2	3	4	5	6	7	8	9			
	g/h	w %	g/h	w %	g/h	w %	g/h	w %	g/h	w %	g/h	w %
Propylene	289	97.0%			257	55.5%	7	3.5%	197	100.0%	53	100.0%
Propane					198	42.7%	198	94.5%				
Hydrogen	9	3.0%	11	100.0%	1	2.5%						
Other hydrocarbons					9	1.8%	4	2.1%			219	100.0%
Oxygen												
Hydrogen peroxide												
Water												
Solvent												
Other oxygen.hydrocarbons												
Propylene oxide												
Total	298	100%	27	100%	11	100%	48	100%	209	100%	197	100%

Component	Stream											
	10	14	12	13	11	15	16	17	18			
	g/h	w %	g/h	w %	g/h	w %	g/h	w %	g/h	w %	g/h	w %
Propylene	160	7.2%					492	65.0%	492	100.0%		
Propane	78	3.5%										
Hydrogen												
Other hydrocarbons												
Oxygen												
Hydrogen peroxide	160	3.6%	64	3.0%	226	5.2%	98	89.9%				
Water	78	3.2%	64	3.0%	4133	94.6%	11	10.1%				
Solvent	2067	89.3%	2067	97.0%	11	0.3%			265	35.0%	265	100.0%
Other oxygen.hydrocarbons												
Propylene oxide												
Total	2305	100%	4435	100%	2131	100%	2131	100%	110	100%	757	100%

CLAIMS

1. An integrated process for the production of olefin oxides which comprises:
 - a) feeding in continuous to a dehydrogenation unit, a paraffinic stream essentially consisting of at least one C₂-C₅ alkyl hydrocarbon;
 - b) sending the stream leaving the dehydrogenation unit to a separation unit to produce a stream consisting of hydrogen, a stream essentially consisting of the non-reacted alkyl hydrocarbon, which is recycled to the dehydrogenation, and a stream essentially consisting of the dehydrogenated alkyl hydrocarbon (olefin stream);
 - c) feeding the stream of hydrogen together with a stream of oxygen, to a hydrogen peroxide synthesis unit;
 - d) feeding a part of the olefin stream, together with the hydrogen peroxide produced in step (c), to an oxidation unit, the rest of the olefin stream being sent to a storage tank;
 - e) recovering the olefin oxide produced and sending it for storage.
2. The process according to claim 1, wherein the hydrocarbon stream is selected from ethane or propane.
3. The process according to claim 1 or 2, wherein the paraffinic stream is fed to the dehydrogenation unit which effects a catalytic dehydrogenation at a temperature rang-

ing from 450 to 800°C and at an absolute pressure ranging from 10 to 300 kPa.

4. The process according to any of the previous claims, wherein the dehydrogenation catalyst is selected from catalysts based on platinum, gallium, chromium, vanadium supported on silica and/or alumina.

5. The process according to any of the previous claims, wherein, at the end of the dehydrogenation, the stream produced is sent to a two-step separation unit.

6. The process according to claim 5, wherein in the first step hydrogen is recovered, while in the second step the non-reacted paraffin is separated from the olefin.

7. The process according to any of the previous claims, wherein the stream of hydrogen and a stream of oxygen are fed in continuous to a stirred reactor containing, in dispersion, a heterogeneous catalyst based on platinum and palladium.

8. The process according to claim 7, wherein a stream is fed to the stirred reactor, consisting of:

- a liquid stream containing an alcohol or an alcohol-water mixture, with an alcohol content higher than 50% by weight, and an acid or halogenated promoter; and
- a gaseous stream containing hydrogen, oxygen and an inert gas, wherein the concentration of hydrogen is lower than 4.5% by volume and the concentration of oxygen is

lower than 21% by volume, the complement to 100 being the inert gas.

9. The process according to any of the previous claims, wherein the hydrogen peroxide synthesis reaction is carried out at a temperature ranging from -10 to 60°C, a pressure ranging from 1 to 300 bars and with residence times in the liquid medium in the reactor ranging from 0.05 to 5 hours.

10. The process according to any of the previous claims, wherein the hydrogen peroxide synthesis catalyst contains palladium in a quantity ranging from 0.1 to 3% by weight and platinum in a quantity ranging from 0.01 to 1% by weight, with an atomic ratio between platinum and palladium ranging from 1/500 to 100/100.

11. The process according to claim 10, wherein the catalytic system comprises one or more promoters selected from metals of group VIII and IB in a concentration not higher than that of the palladium.

12. The process according to any of the previous claims, wherein the hydrogen peroxide synthesis catalyst is dispersed in the reaction medium at a concentration ranging from 0.1 to 10% by weight.

13. The process according to claim 8, wherein the liquid stream consists of one or more C₁-C₄ alcohols or of a mixture of said alcohols with water.

14. The process according to claim 8, wherein the acid

promoter is selected from sulfuric, nitric and phosphoric acids or from sulfonic acids.

15. The process according to claim 14, wherein the concentration of the acid generally ranges from 20 to 1000 mg per kg of liquid medium.

16. The process according to claim 8, wherein the halogenated promoter consists of a substance capable of generating halogen ions in the liquid reaction medium.

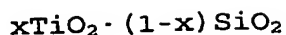
17. The process according to claim 16, wherein the concentration of halogenated promoter generally ranges from 0.1 to 50 mg per kg of liquid medium.

18. The process according to any of the previous claims, wherein the olefin stream and hydrogen peroxide are fed to the epoxidation reactor under such conditions as to have a molar ratio olefin/H₂O₂ inside the reaction container ranging from 10/1 to 1/10.

19. The process according to any of the previous claims, wherein the epoxidation reaction is carried out under stirring, in the presence of the same solvent used in the hydrogen peroxide synthesis, at a pH ranging from 5.5 to 8 and in the presence of a catalyst used in a quantity ranging from 1 to 15% by weight with respect to the reaction mixture.

20. The process according to any of the previous claims, wherein the epoxidation catalyst is titanium-silicalite

having the general formula:



wherein x represents a number ranging from 0.0001 to 0.04, preferably from 0.01 to 0.025.

- 5 21. The process according to any of the previous claims, wherein the epoxidation reaction of the olefin is effected at a temperature ranging from 20 to 150°C and at such a pressure as to keep the olefin dissolved in liquid phase at the reaction temperature.
- 10 22. The process according to any of the previous claims, wherein the stream leaving the epoxidation reactor is sent to a purification section from which the olefin oxide produced is recovered and sent for storage, together with the solvent which is recycled both to the hydrogen peroxide
- 15 synthesis reactor and to the oxidation reactor of the olefin.
23. An integrated process for the production of olefin oxides which comprises:
- a) feeding in continuous to a dehydrogenation unit, a par-
- 20 affinic stream essentially consisting of at least one C₂-C₅ alkyl hydrocarbon;
- b) sending the stream leaving the dehydrogenation unit to a first separation unit to produce a stream consisting of hydrogen and a mixed stream essentially consisting of
- 25 the non-reacted alkyl hydrocarbon and the dehydrogenated

alkyl hydrocarbon (olefin);

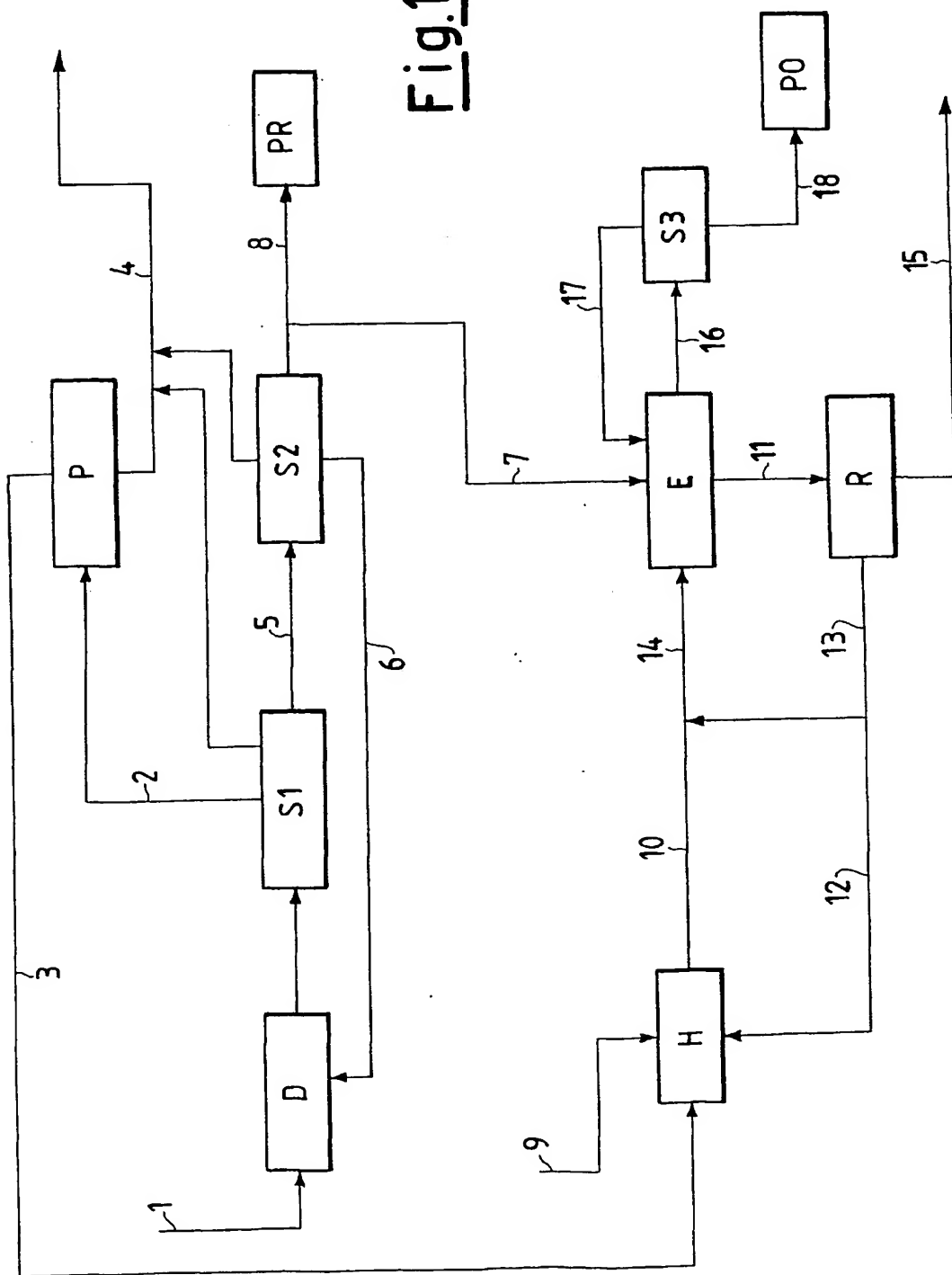
c) feeding the stream of hydrogen together with a stream of oxygen, to a hydrogen peroxide synthesis unit;

d) feeding a part of the hydrocarbon stream coming from the first separation section, together with the hydrogen peroxide produced in step (c), to an oxidation unit;

e) feeding the stream leaving the epoxidation unit to a second separation unit to recover a stream containing the olefin oxide produced and a stream essentially consisting of the non-dehydrogenated alkyl hydrocarbon which is recycled to the dehydrogenation;

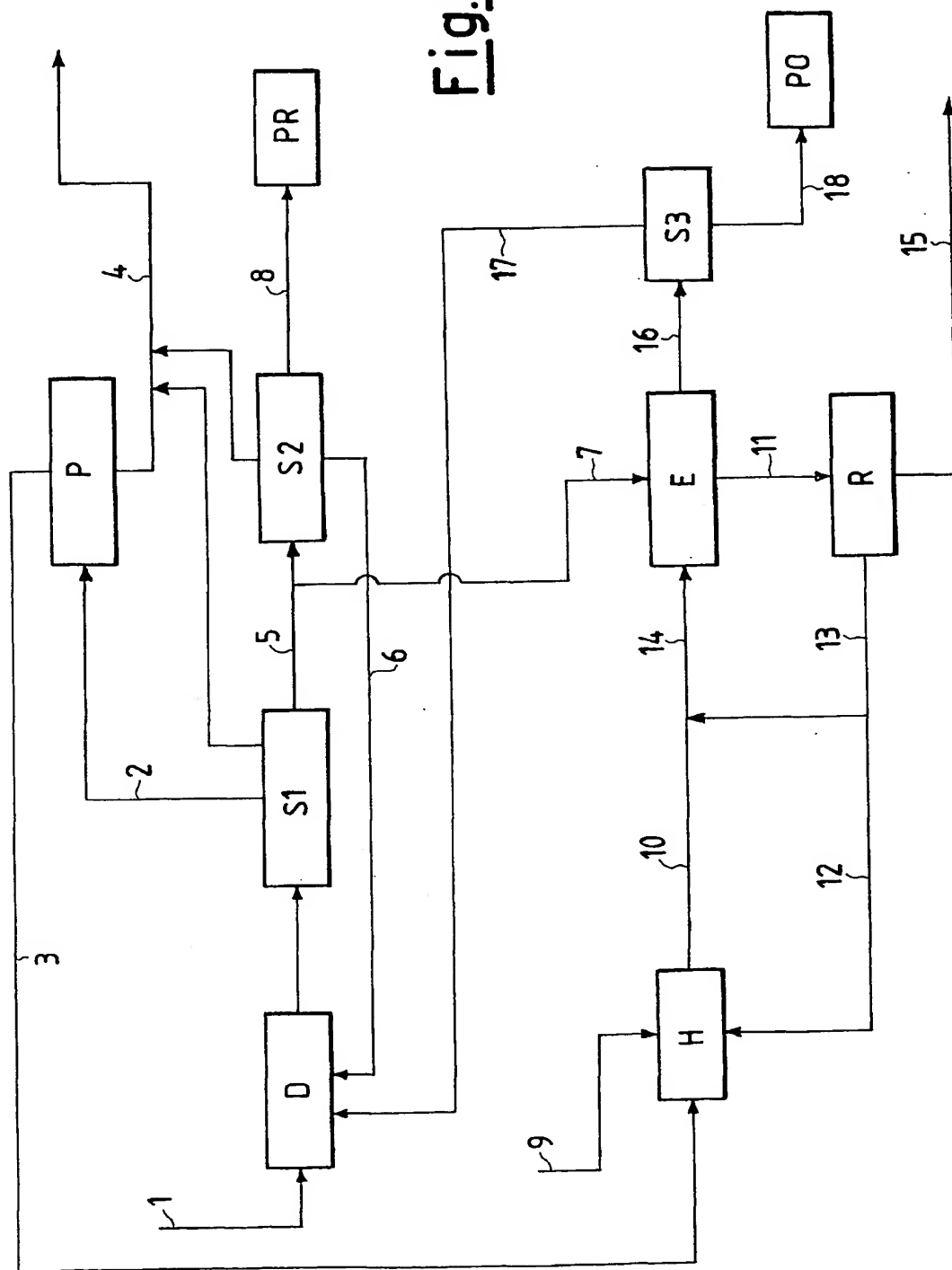
f) feeding the remaining part of the hydrocarbon stream coming from the first separation section to a third separation section to recover a paraffin stream, recycled to the dehydrogenation, and an olefin stream sent for storage.

Fig.1



2/2

Fig.2



INTERNATIONAL SEARCH REPORT

International Application No

PCT/EP 02/03299

A. CLASSIFICATION OF SUBJECT MATTER
IPC 7 C07D301/12

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 7 C07D

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

CHEM ABS Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 5 599 956 A (PETER R. PUJADO) 4 February 1997 (1997-02-04) the whole document	1-23
A	WO 00 20404 A (ARCO CHEMICAL TECHNOLOGY) 13 April 2000 (2000-04-13) the whole document	1-13

☐

Further documents are listed in the continuation of box C.

☒

Patent family members are listed in annex.

* Special categories of cited documents:

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Date of the actual completion of the international search

9 August 2002

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INTERNATIONAL SEARCH REPORT

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International Application No

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Patent document cited in search report		Publication date	Patent family member(s)	Publication date
US 5599956	A	04-02-1997	NONE	
WO 0020404	A	13-04-2000	US 5973171 A AU 6201499 A WO 0020404 A1	26-10-1999 26-04-2000 13-04-2000